

Probability model of solid to liquid-like transition of a fluid suspension after a shear flow onset

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Abstract

The complexity of interactions of different origin between particles in dense fluid suspensions limits application of the concepts of classical mechanics for finding a relation between individual particles dynamics, and the macroscopic dynamic response. For this reason, the probability model of suspension flow after a shear flow onset is proposed that incorporates our certain lack of understanding of environments effects. These effects are identified in the model with the random rate of particles arrival to flow. The multiplication rule is used to sweep from the individual particles to the entire system. The resulting probability of the system shift to flow represents well the measured boundary position, separating the flowing suspension from its solid-like state, and proves thus the appropriate choice of representation of the particle interaction effects.

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1. Introduction

The recent reviews by Barnes (1997, 1999) recapitulate a large amount of studies on dense fluid suspensions. The prevailing number of the cited papers deals with the mechanical response of suspensions to stressing or straining mechanism. Nevertheless, the attention is paid also to the flow problems and the consequent role of the interface formation between the flowing and the stationary section of suspension. The origin of the two sections separated by the interface depends on the proportion of acting hydrodynamic forces and inter-particle attractive forces. When the hydrodynamic disturbances predominate the attractive forces between particles the fluid suspension flows and vice versa.

We have recently monitored the interface propagation in the fluid suspension confined between two coaxial cylinders by the ultrasound waves and visualization techniques con-

sidered in Nouar and Riha (2000) and Nouar et al. (2003). Our experimental results, which are summarized in the next section, help to clarify the progress of individual particles transiting from solid to fluid flow states and their participation into the response of the entire system to mechanical loads. The displacement of the interface between the flowing and stationary regions has also been observed recently by Raynaud et al. (2002) in relation to the measurement of velocity distribution in a fluid suspension, using the magnetic resonance imaging technique. The displacement of the asymptotic interface position determined from the velocity profiles was found to dictate the apparent suspension flow behavior.

A relation between the microscopic individual particles dynamics and their macroscopic response can be found, for instance, only in the case of dilute suspensions (Larson, 1999). In case of concentrated suspensions, the description cannot be accurate owing to the potential inaccuracies resulting from the uncertainty of inter-particle hydrodynamic, electrostatic, van der Waals, etc., interactions. The lack of knowledge of an inner flow mechanism encourages

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us to translate the result of our experimental observation into the probabilistic scheme. Within the framework of this scheme, the finding of gradual particle transition to flow is used to represent the particle arrival into the liquid-like section and to specify the interface propagation. The resulting probability function is used to determine the experimentally measured interface approach to the equilibrium position and, consequently, to justify the assumed effect of particles interaction on the mechanism of transition to flow.

2. Experimental results

The fluid used for the experiments is the suspension of industrial bentonite (particle size $<2\ \mu\text{m}$, particle concentration 6 wt%, i.e., 2.3 vol%) in a deionised water solution of carboxymethyl-cellulose (0.5 wt%). The suspension is prepared by agitation of compounds for 2 h, filtration and suspension structure formation at rest during 1 week.

The time necessary for a complete suspension restructuring is determined using the Weissenberg Rheogoniometer with the cone plate system (6 cm, 2°). Several start-ups preshearing are successively undertaken with different periods of rest between each of them at the shear rate $10\ \text{s}^{-1}$. The change in the material response is found insignificant after the rest period of 6 h. The suspension was protected against water evaporation by covering the free suspension surface by dodecane ($\text{C}_{12}\text{H}_{26}$, mass density $0.7\ \text{kg}/\text{dm}^3$).

The experimental set-up for the measurement of interface propagation consists of two coaxial cylinders. The unit of the controlled stress viscometer rotates the inner metal cylinder what makes possible to monitor the inner cylinder velocity. The outer cylinder is machined from a cubic plexiglas bloc that is connected and centralized with the fixed part of the viscometer. The radius of the inner and outer cylinder is 20 and 50 mm, respectively. The height of inner cylinder submerged in the suspension is 55 mm. To avoid the suspension slip on the cylinder surfaces, the inner and outer cylinder surfaces are roughened by a glued emery cloth with the equivalent roughness of $100\ \mu\text{m}$.

The suspension is kept at rest for 6 h in the gap between cylinders before the start of each flow experiment. All experiments are carried out at the ambient temperature $20 \pm 2\ ^\circ\text{C}$. An additional precaution is taken to prevent the loss of moisture to the atmosphere by covering the suspension surface with a transparent plastic film sealed with the plexiglas cube. The plastic film is removed during the visualization tests. It is believed that the water evaporation is negligible in the course of the test, that is, during 1200 s.

The ultrasonic velocity profile monitor (Met-Flow, model X-2) is used for the measurement of tangential velocity distribution in 30 mm gap between cylinders. The schematic of the cross-section of the experimental set-up and the position of the ultrasound transducer is shown in Fig. 1. The position of the transducer ensures that the cylindrical ultrasonic beam to which the acoustic energy is supposedly concentrated is tangential to the inner cylin-

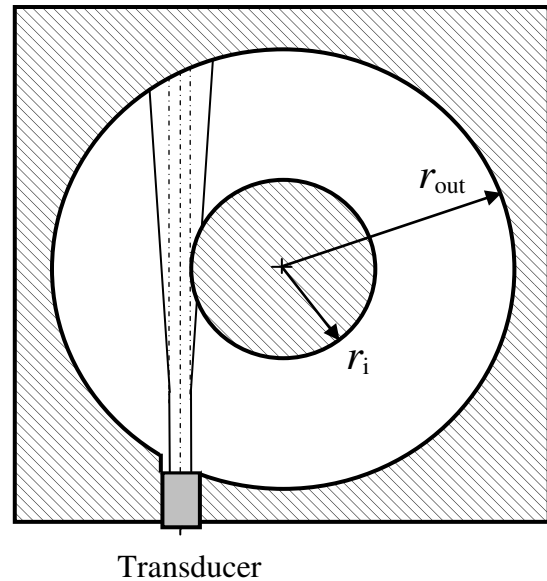


Fig. 1. The schematic showing the cross-section of the experimental set-up for the measurement of the velocity distribution between two concentric cylinders with radii $r_i = 20\ \text{mm}$ and $r_{\text{out}} = 50\ \text{mm}$, the position of the ultrasound transducer and the shape of the ultrasonic beam.

der as indicated by the dashed lines in the figure. The effective diameter of the beam is equal to the diameter of the piezoelectric element emitting the ultrasonic pulses.

When suspension is subjected to strain by the imposed constant torque to the inner cylinder, which does not break up completely the particle network structure across the gap, the straining causes elastic, then plastic and finally a viscous suspension deformation at a particular position. The elastic deformation increases up to the elastic-limit strain ($\gamma_e = 0.065$, dimensionless) defined as the maximum recoverable strain at the imposed load. A subsequent plastic deformation (creep) advances up to the corresponding plastic-limit strain ($\gamma_g = 0.4$) defined as the strain limit at which the sudden strain increase owing to the viscous deformation is observed. Both strain limits were determined by means of the controlled stress rheometer AR 1000. When suspension deformation is higher than the plastic-limit strain and begins to increase abruptly, the particles are released from the inter-particle bonding, at the surface of the stationary section, and started to flow forming the liquid-like section between the interface and the inner cylinder. The interface radial motion to the equilibrium position, after the onset of shear strain, was monitored by the visualization technique. A narrow stripe of powder (TiO_2) was spread in the radial direction on the suspension free surface between the cylinders before the experiment. The representative superposition of the tracer stripes is shown in Fig. 2. The stripe distortion was recorded by CCD camera at the speed 25 images/s. The recorded images are then digitized into 512×512 pixels and analyzed using Matlab software. Afterwards, the angular displacement $\alpha(r)$ of the stripe (the displacement between the initial angular position on the strip at radius

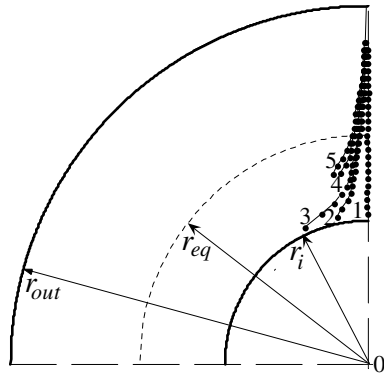


Fig. 2. The superposition of the stripes of tracer on the suspension free surface observed at the time $t = 0$ s (curve 1), $t = 1$ s (curve 2), $t = 5$ s (curve 3), $t = 21$ s (curve 4) and $t = 1200$ s (curve 5). The applied torque at the inner cylinder is 28.3×10^{-4} Nm. The corresponding shear stress is 20.45 Pa.

r and the angular position at particular times) is evaluated. The angular displacement and the radial position define the local shear strain as $\gamma(r) = r|d\alpha/dr|$.

The normalized strain evolution $(\gamma_g - \gamma)/\gamma_g$ at a given radial position, before the plastic-limit strain γ_g is achieved, is shown in Fig. 3 for three dimensionless radial positions $(r_{eq} - r)/(r_{eq} - r_i) = 1/3, 1/2$ and $2/3$. In our experiment (see Nouar et al., 2003), the radius of the inner cylinder $r_i = 20$ mm and the equilibrium position of the interface is located at $r_{eq} = 32$ mm. The presented typical data indicate that the particle transition to flow proceeds successively. The particles (or the particle clusters) near the driven inner cylinder start to flow prior to the particles located far away. Finally, the last particles, which start to flow, are located near the equilibrium interface position.

The typical details of strain evolution in the stationary section are shown in Fig. 4. The fine time scale reveals their

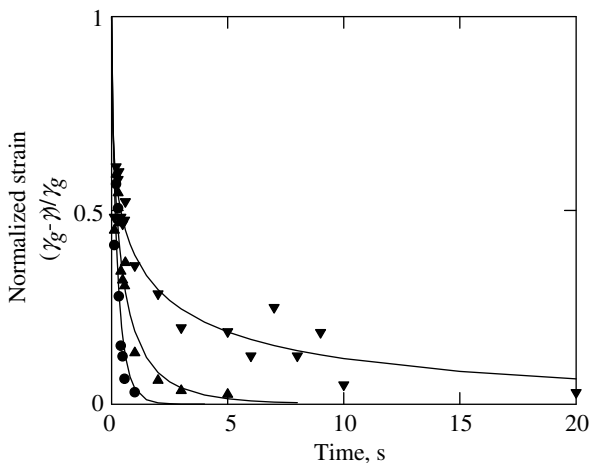


Fig. 3. The normalized shear strain at different non-dimensional radial positions between the inner cylinder and the interface equilibrium position. The full circles, the full triangles and the full down pointing triangles correspond to the non-dimensional radial position 1/3, 1/2 and 2/3, respectively. The full exponential lines help to guide the eyes.

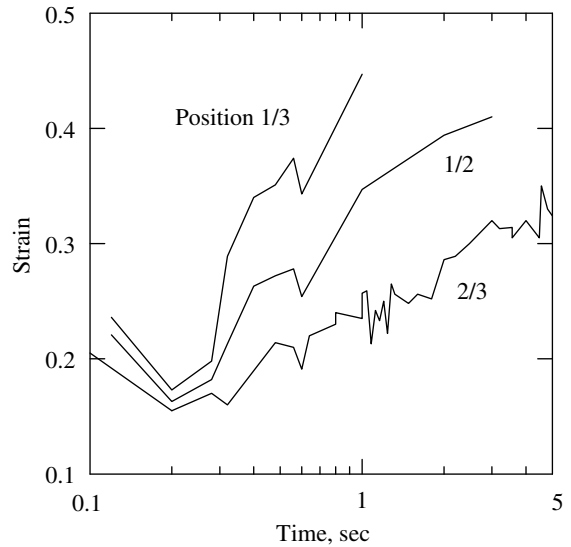


Fig. 4. The shear strain evolution at the non-dimensional radial positions 1/3, 1/2 and 2/3 between the inner cylinder and the equilibrium position of interface, respectively. The strain fluctuations stem presumably from the acting of varying hydrodynamic shear stress on the interface and the elastic contractions of the solid-like suspension section.

fluctuations illustrated by lines which connect the measured values of local strain evolution at intervals about 100 ms. We believe that these fluctuations stem from the acting of varying hydrodynamic shear stress on the interface surface. The explanation of this mechanism leading to the stress variation, thus the strain fluctuations, may be presented as follows. Owing to the imposed constant torque on the inner cylinder at the start-up of the suspension straining, the shear stress acting on the interface surface increases, which increases the monitored strain in the stationary suspension section. When a critical strain value, γ_g , is reached, the particle or the particle cluster, in some axial position along the interface surface, is torn off from the stationary suspension. Consequently, the local interface position suddenly shifts towards the outer cylinder that increases the width of the liquid-like section. As a result, the stress decreases at the interface surface and, consequently, the strain decreases also in the solid-like section owing to the elastic contraction of the solid-like suspension. Afterward, the stress starts again to increase as well as the strain at the interface until it reaches the plastic-limit, γ_g , then the cycle manifesting strain oscillations is repeated again.

The above plausible reasoning about the gradual particle transition to flow is used in the following section to propose the probability description of this transition. This description will link the effect of individual particles on the bulk behavior of the fluid suspension, which is monitored by the interface propagation after the onset of shear flow.

3. Probability description of the onset of flow

The particles transition to flow is a deterministic physical process caused by the onset of shearing of the

stationary solid-like suspension by the inner cylinder. The formation of the solid-like suspension state is completed after sufficient period of time (6 h) before the start of each flow experiment. When the local strain after the onset of inner cylinder rotation is higher than the plastic-limit strain ($\gamma_g = 0.4$), the particles (or the particles aggregates) are released from the inter-particle bonding, at the surface of the stationary section, and start to flow forming the liquid-like section between the interface and the inner cylinder. The events when the respective units of one or more particles leave the solid-like section of the suspension and arrive into the flowing liquid-like section are time dependent. The first event (unit arrival) occurs after a certain amount of time spent from the beginning at some point of time. Then, after a certain amount of time, the second event occurs. The time between consecutive events (inter-arrival interval) is certainly irregular. We cannot predict exactly when an event will occur because of the limitations to our knowledge of the transition process. Consequently, we resort to the probability description of particles arrival into the liquid-like section that incorporates our uncertainty assuming randomness of inter-arrival intervals.

Owing to the possible particles binding, the number of arrivals (events) is not a deterministic but rather an integer-valued random variable N ($N \leq m$), where m represents the total number of individual particles taking part either separately or in aggregates in the whole transition process. The capital letter is used here to denote the random variable when the lower-case letter denotes non-random one.

The inter-arrival intervals T_n form a sequence T_1, \dots, T_N , where T_1 denotes time until the first event, T_n time between the $(n-1)$ st and the n th event for $n > 1$, and T_N time between the $(N-1)$ st and the last N th event (the last particle or particles unit arrival). The random inter-arrival intervals $\{T_n, n = 1, 2, \dots, N\}$ are by definition disjoint.

The probability $\Pr(T_n > t)$ of an inter-arrival interval T_n being greater than time t will be represented by a continuous-time model $\Pr(T_n > t) = \exp(-\lambda(t))$ with a non-constant rate function $\lambda(t)$. Obviously, the probability can be easily calculated when the rate function is limited to the constant value, $\lambda = \text{constant}$, which results in an exponential decrease, i.e. $\Pr(T_n > t) = \exp(-\lambda t)$. This limit is a particular case when any interactions of different origin between particle units are neglected (Berlin et al., 1993). However, the constant λ value approximation is too crude to represent the probability of an inter-arrival interval for a dense fluid suspension when the environment effects on the particles transition are involved. These effects will be identified in the continuous-time model by the random arrival rate λ_n such that the probability $\Pr(T_n > t)$ is conditioned by the value λ_n taken by λ_n ,

$$\Pr(T_n > t | \lambda_n) = \exp(-\lambda_n t). \quad (1)$$

The rate λ_n reflects the correlated particle arrival dynamics owing to the possible inter-particle interactions in dense suspensions. Each particle is locked at rest into an interac-

tion state which owing to imposed particular local strain can result a set of possible inter-arrival intervals due to the possible different arrival rates λ_n . The conditional probability $\Pr(T_n > t | \lambda_n) = \exp(-\lambda_n t)$ corresponds to one of the possible inter-arrival interval. According to the total probability theorem, the probability $\Pr(T_n > t)$ may be expressed by means of the expected value of the conditional probability as,

$$\Pr(T_n > t) = E[\Pr(T_n > t | \lambda_n)] = E[\exp(-\lambda_n t)], \quad (2)$$

where E stands for the expected value operator.

The random inter-arrival intervals $\{T_n, n = 1, 2, \dots, N\}$ and the random rates $\{\lambda_n, n = 1, 2, \dots, N\}$ are considered to form sequences of independent and identically distributed random variables. This assumption may not completely hold for dense fluid suspensions. However, given a certain lack of understanding of the mechanism that controls particles arrival, the independence assumption is legitimate and leads to the first-approximation model in attempting to model the units arrival into the liquid-like section.

The joint probability $\Pr(T > t)$ that is the probability of the joint occurrence of independent events follows the multiplication rule

$$\begin{aligned} \Pr(T > t) &= E[\exp(-\bar{\lambda}_N t)] = \prod_{n=1}^N \Pr(T_n > t) \\ &= \prod_{n=1}^N E[\exp(-\lambda_n t)]. \end{aligned} \quad (3)$$

T denotes the effective arrival interval of entire system of N units into the liquid-like section and the effective arrival rate $\bar{\lambda}_N$ represents a contribution of individual particles rates to this interval. The probability $\Pr(T > t)$ represents in these circumstances the probability that the arrival interval of the entire system of N units is greater than time t .

The effective rate $\bar{\lambda}_N$ incorporates contributions of individual rates since $\bar{\lambda}_N = \sum_{n=1}^N \lambda_n$. Moreover, when the system consists of a large number of units, then according to the central limit theorem, the distribution of $\bar{\lambda}_N$ can be satisfactorily approximated by the weak limit of λ_n . When suitably normalized, the weak limit A depends only on the probability distribution associated with the λ_n , $A = \lim_{N \rightarrow \infty} \bar{\lambda}_N = \lim_{N \rightarrow \infty} \frac{1}{a_N} \sum_{n=1}^N \lambda_n$, where $a_N > 0$ denotes a sequence of suitable normalizing constants (Jurlewicz and Weron, 1999, 2002; Hetman et al., 2003; Jonscher et al., 2003). The limit A can represent the effective rate in Eq. (3) instead of $\bar{\lambda}_N$. The necessary and sufficient condition for the convergence of the sum is that the distribution of λ_n belongs to the domain of attraction of the one-sided Lévy stable distribution of A , $A \stackrel{d}{\cong} \tau S_\alpha$. The sign $\stackrel{d}{\cong}$ denotes the equality in distribution, the constant τ is positive, α the parameter ($0 < \alpha < 1$) and S_α is such a random variable (Lévy stable non-negative random variable) that its Laplace transform is the stretched exponential function (Jurlewicz and Weron, 2000; Hetman et al., 2003)

$$E[\exp(-sS_x)] = \int_0^\infty \exp(-st)f_x(t)dt = \exp[-s^x], \quad (4)$$

where $f_x(t)$ represents the probability density function of S_x . Hence, it follows from (4) that the probability $\Pr(T > t)$ of the effective inter-arrival interval T being greater than time t is

$$\Pr(T > t) = E[\exp(-At)] = \exp[-(\tau t)^x]. \quad (5)$$

The probability function $\varphi(t) = \Pr(T > t)$ is stretched exponential with the initial value $\varphi(0) = 1$ and the limit $\varphi(t) \rightarrow 0$ as $t \rightarrow \infty$.

As stated above, the inter-arrival intervals $\{T_n, n = 1, 2, \dots, N\}$ are considered independent and identically distributed random variables. Consequently, with respect to the considered probability model, the event (the particle arrival into the liquid-like section) in one interval is independent of the event in any other interval. Moreover, the events do not occur simultaneously, since we consider arrivals consisting of individual particle or conjoint particles as one event. This is expressed by the probability of the events in time Δt

$$\lim_{\Delta t \rightarrow 0} \Pr(X(t + \Delta t) - X(t) > 1 | X(t + \Delta t) - X(t) \geq 1) = 0, \quad (6)$$

where a non-negative integer-valued random variable $X(t)$ denotes the number of events (the number of particle units arriving into the liquid-like section) in the course of time $[0, t]$.

The inter-arrival interval T_1 of the first event is more than t if the number of events $X(t)$ before time t is a 0. Consequently, the probabilities for this event are the same $\Pr(T_1 > t) = \Pr(X(t) = 0)$. Similarly, the effective inter-arrival interval T of the first event (the arrival of the entire system of N units into the flowing section) is more than t if there is no event (the arrival of the entire system of N units) before time t .

The deterministic arrival process has besides the counting aspect, that is, how many particles units arrived into the liquid-like section, also a geometrical one. The particles have a size and thus their cumulation enlarges the volume of this section. The experimental observation shows that the particles unit near the driven inner cylinder starts to flow prior to the unit far away (see Figs. 2 and 3). Consequently, the position r_n of the flowing n th unit in the liquid-like section, which is adjacent to the stationary particle in the stationary section, may be associated with the local position, r , of the interface at time, t . The radial position, r_n , of flowing particle unit remains fixed when there is no additional diffusive, thermal diffusive and/or convective particle flow to the circumferential Couette flow between two cylinders induced by the rotation of the inner one. This view holds for the isothermal flow in the liquid-like section but it is not necessarily true for the possible thin interfacial layer between the flow and the stationary sections. For this reason, the interfacial layer, where the particle transition to

the flow may not be completed, is considered to belong to the stationary section.

Within the framework of the chosen two sections concept, the particle release from the inter-particles links in the stationary section ensures an equivalent particle arrival into the liquid-like section. Thus the interface position, $r(t)$, is proportional to the number of units $X(t)$ in the liquid-like section. When $X = 0$, then $r = r_i$, where r_i is the radius of the inner cylinder. On the other hand, when $X = N$, the position of the last N th unit determines the interface equilibrium position r_{eq} , $r_{eq} = r_N$.

As far as the interface shape is concerned, two sections model implies that the radial position r_n of n th flowing particle unit forms a cylindrical boundary of the flowing suspension portion, which consists of n particle units. When the particle location r_n localizes the interface position, the interface between the stationary and the liquid-like sections, at position $r(t) = r_n$, is also cylindrical.

The successive arrival of N particles units into the liquid-like section means the interface propagation between its initial position at time $t = 0$ at the inner cylinder, $r = r_i$, and its final position, $r = r_{eq}$ as $t \rightarrow \infty$. The tendency of interface shift from the initial position $r = r_i$ to the position $r = r_1$ is bound with the probability $\Pr(T_1 > t) = \Pr(X(t) = 0)$, that is, the probability of the first unit arrival into the liquid-like section. Accordingly, the tendency of interface shift between the position $r = r_i$, and its final position, $r = r_{eq}$, is related to the probability $\Pr(T > t)$ that the effective inter-arrival interval T for the equivalent aggregate representing the entire system of N units is greater than time t .

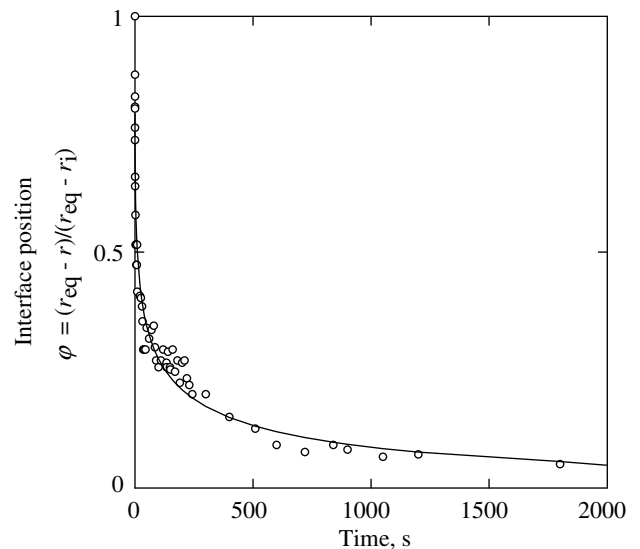


Fig. 5. The typical evolution of the non-dimensional interface position between two coaxial cylinders after the start-up flow induced by the imposed torque (28.3×10^{-4} Nm) on the inner cylinder. The equilibrium interface position $r_{eq}/r_{out} = 0.64$. The symbols denote the experimentally measured positions of the interface and the solid line represents the prediction by means of the probability function Eq. (7).

The deterministic macroscopic process consisting of many mesoscopic arrival events was observed experimentally as the interface move between the initial position $r = r_i$, and its final position, $r = r_{eq}$, Fig. 5. The tendency of this move was mathematically constructed and results in the main outcome of this work, namely, the probability $\Pr(T > t)$ of the effective inter-arrival interval T being greater than time t and the probabilistic function $\varphi(t) = \Pr(T > t)$, Eq. (5). The following relation of the probability function to the interface position links appropriately the model prediction with the observed interface propagation,

$$\varphi(t) = (r_{eq} - r)/(r_{eq} - r_i) = \exp[-(\tau t)^\alpha], \quad (7)$$

where $r(t)$ stands for a certain radial interface position, $\varphi(0) = 1$ and $\varphi(t) \rightarrow 0$ as $t \rightarrow \infty$. The typical data presented in Fig. 5 are very well represented when the parameters are: $\tau = 0.025$ and $\alpha = 0.28$. The corresponding equilibrium interface position is located at $r_{eq} = 32$ mm. This value follows from the balance of momentum for a steady shear flow (Nouar et al., 2003).

4. Final remarks

The primary aim of this paper is to propose a model of suspension particles transition to flow after the onset of shear flow. This aim came up after our observation of the fluid suspension particles transition to flow between coaxial cylinders (Nouar and Riha, 2000; Nouar et al., 2003). The mechanism of individual suspension particle transition to flow has not been studied yet in details as pointed out in this paper. Though, the industrial use of fluid suspension as well as the investigation of their flows and flow properties is very extensive (Barnes, 1997, 1999).

The suspension particles transition to flow is a deterministic physical process caused by the onset of shearing of the stationary fluid suspension. The process involves the recurrence of events in time when the respective units, consisting of one or more particles, join the flowing portion of the suspension.

Owing to the deficiency of our knowledge of the deterministic transition process when every event has a cause, the probability description of events incorporates our uncertainty. To propose at least the first-approximation model, the interaction of particles was included into the probabilistic model in a very simple way, namely, through the randomness and independence of the units arrival rates A_n . The following mathematical construction links the contribution of individual particle units with rates A_n to the characteristic rate Λ for the entire system. The only possible probability distribution for Λ transforms the stochastic system into the deterministic representation, Eq. (5), which manifests the stretched exponential response to the induced units arrival to the flowing section. The reasonably good description of the measured interface position by the predictive relation (7) shown in Fig. 5, justifies our probability

model for the start-up of flow of a fluid suspension with included effect of particles interaction.

A traditional way to describe a stretched exponential data as shown in Fig. 5, is by using the weighted summation of a number of independent exponential functions. However, this procedure is a formal mathematical tool and does not clarify the mechanism of individual suspension particles transition to flow. On the other hand, the probabilistic approach discloses possible internal features of the transition process, which govern the global stretched exponential behavior. These features are generated by the random inter-arrival intervals of the particle arrivals into the flowing section and by the form of the characteristic arrival rate Λ for the entire system.

The investigation is a contribution to studies on the so-called yielding behavior of suspensions (Barnes, 1997, 1999) and to industrial problems associated with, e.g., the start-up of pipeline transporting waxy crude oil (Chang et al., 1999), etc.

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